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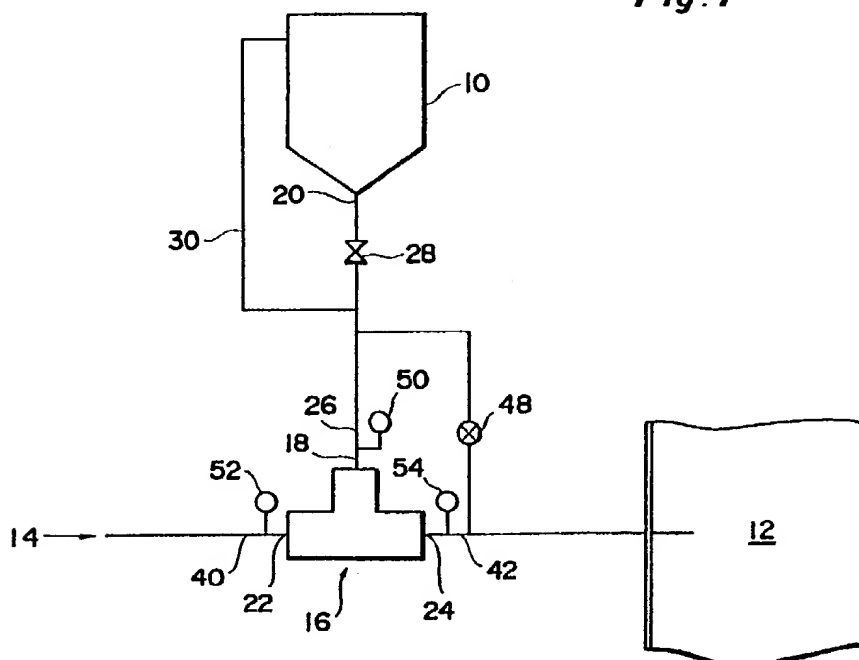
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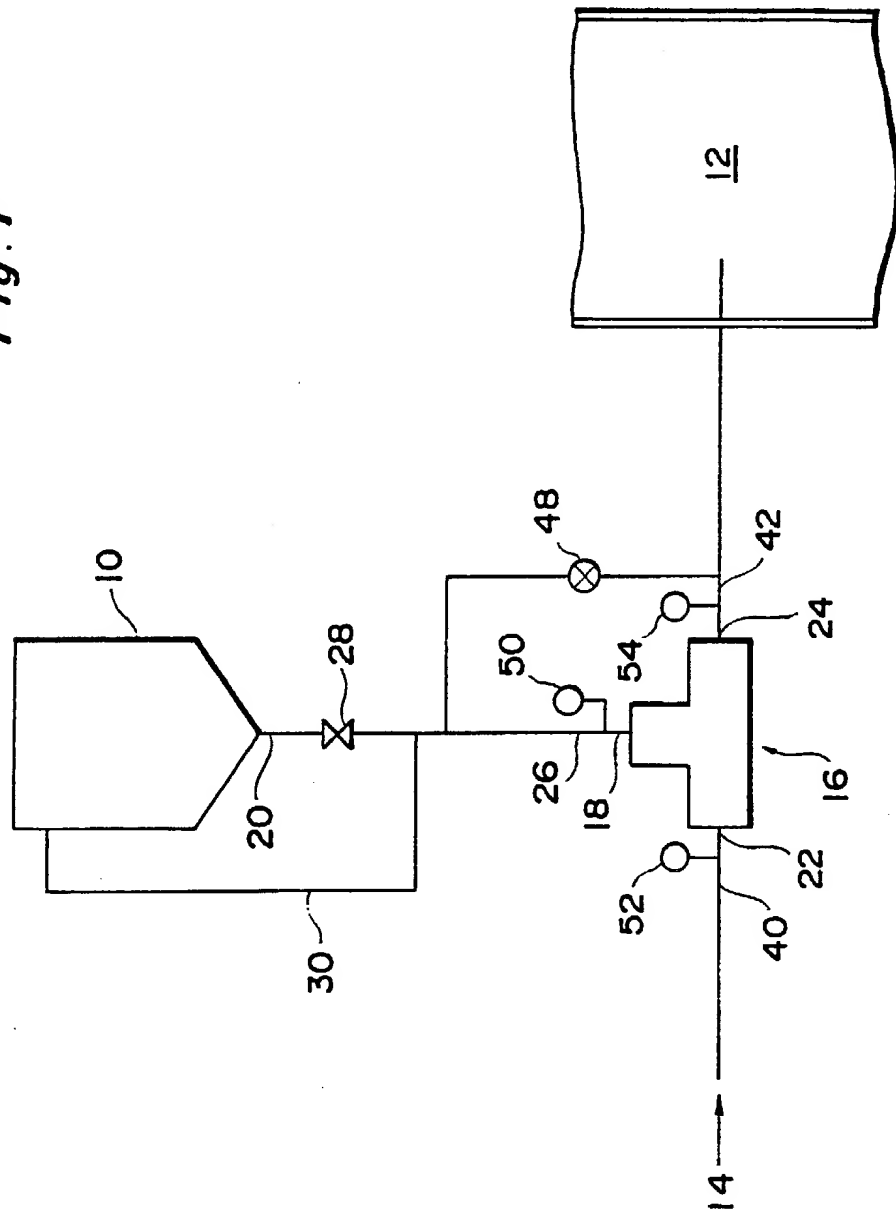
## (54) Supplying catalyst for a gas-phase polymerisation

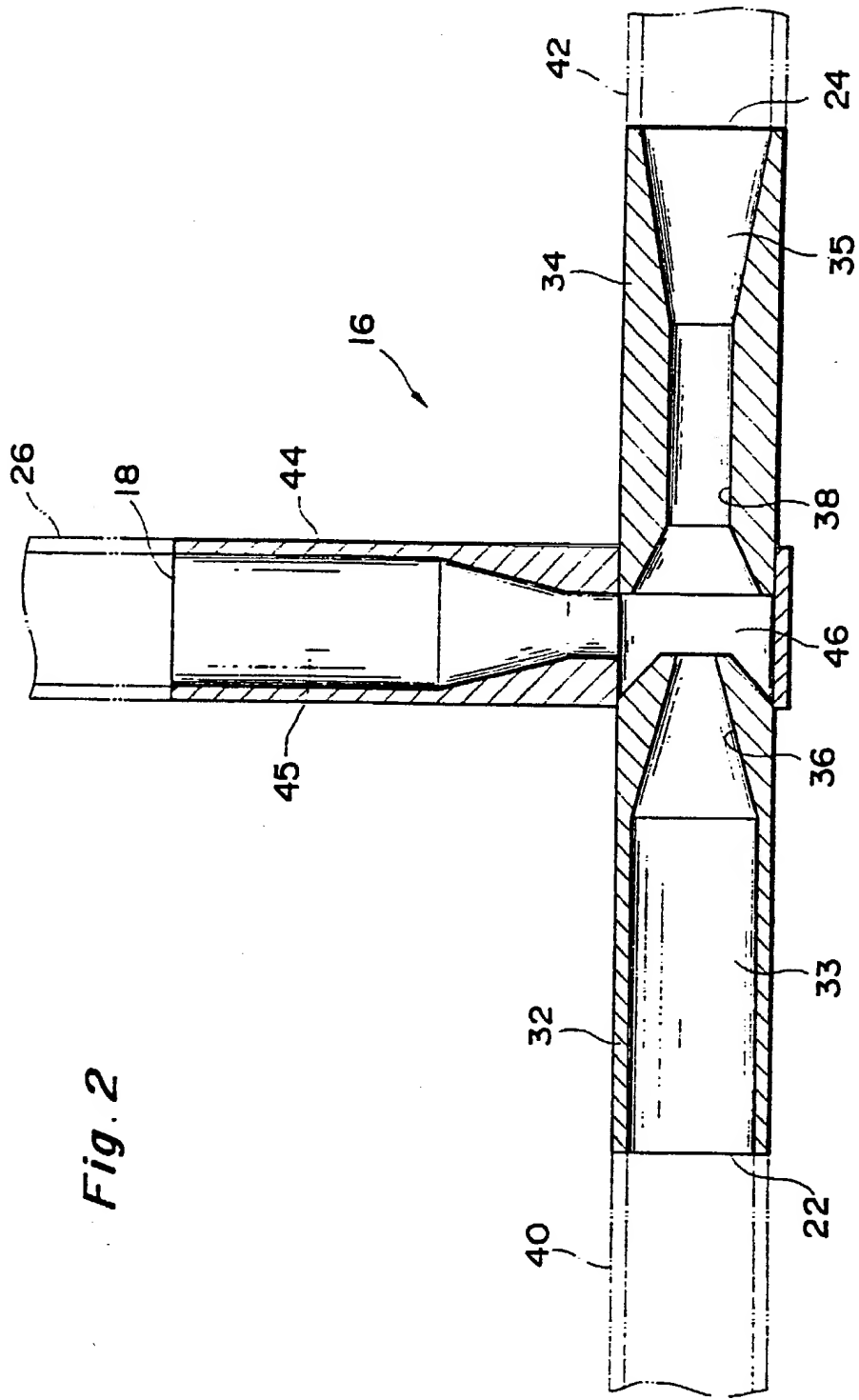
(57) A system for supplying a powder catalyst to a gas phase polymerization reactor together with a gas serving as a polymerization material includes a catalyst storage container (10), a gas supply source (14), and a jet pump type powder supply apparatus. The catalyst storage container can store and supply the powder catalyst. The gas supply source supplies the polymerization material gas. The jet pump type powder supply apparatus (16) generates a negative pressure therein by accelerating a flow of the gas flowing from the gas supply source, draws the powder catalyst from the catalyst storage container by the negative pressure, and supplies the powder catalyst to the gas phase polymerization reactor together with the gas.

Fig. 1



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*Fig. 1*

*Fig. 2*

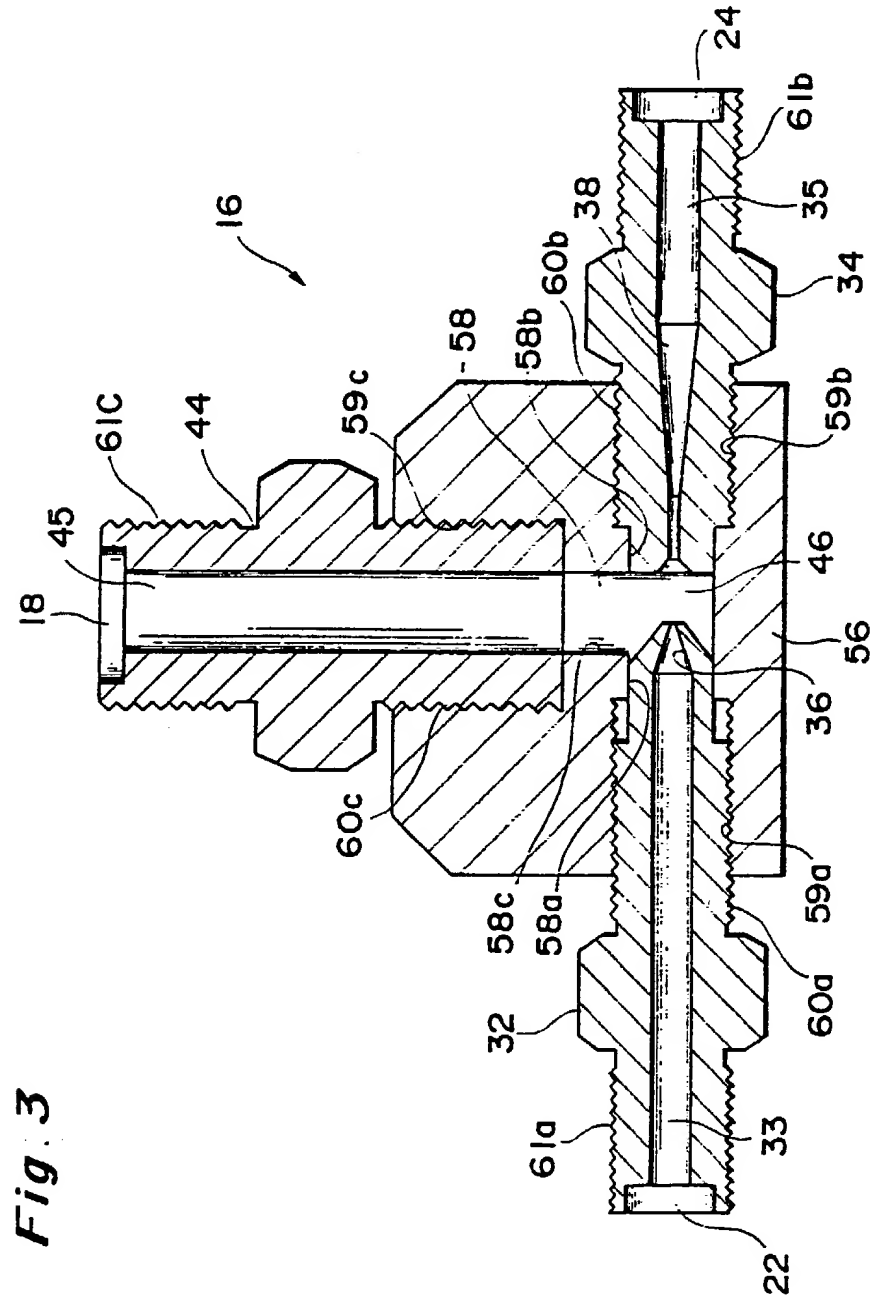


Fig. 4

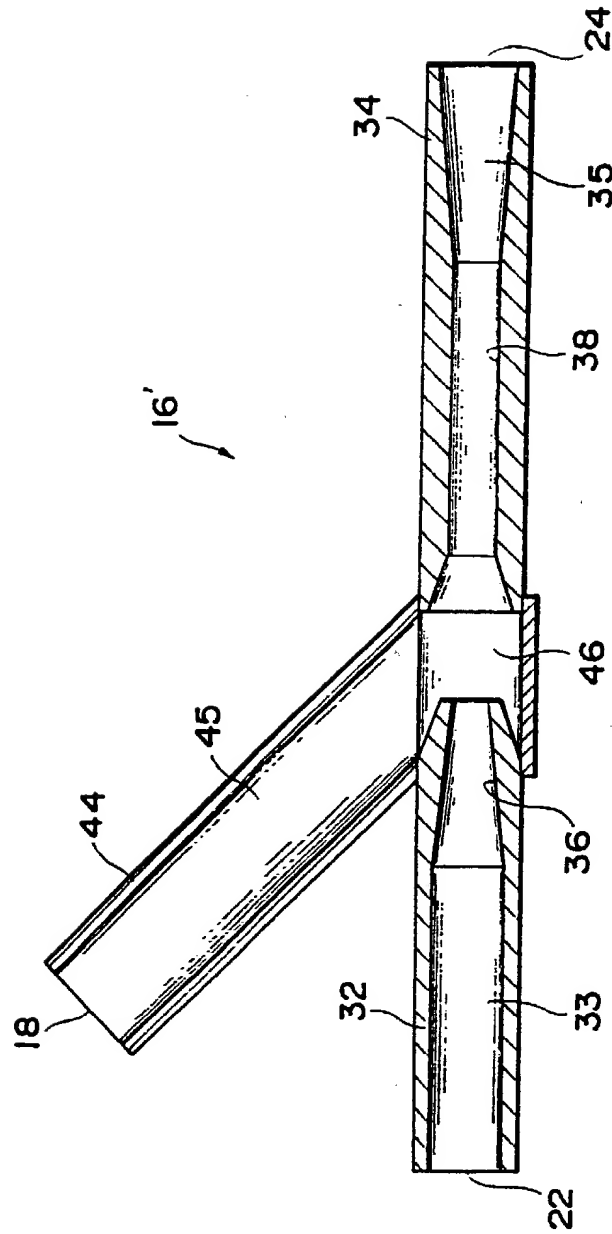


Fig. 6

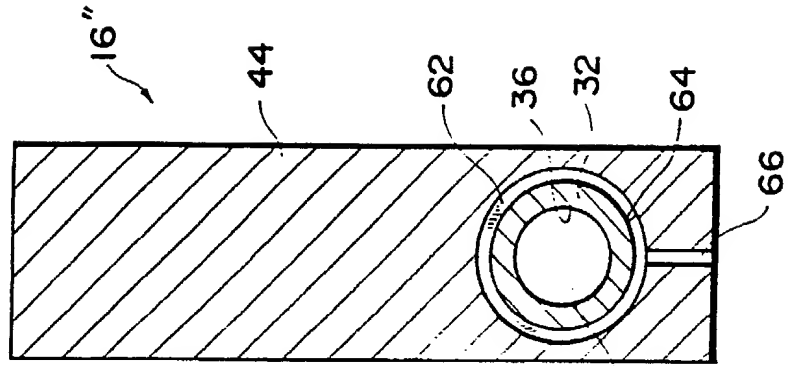
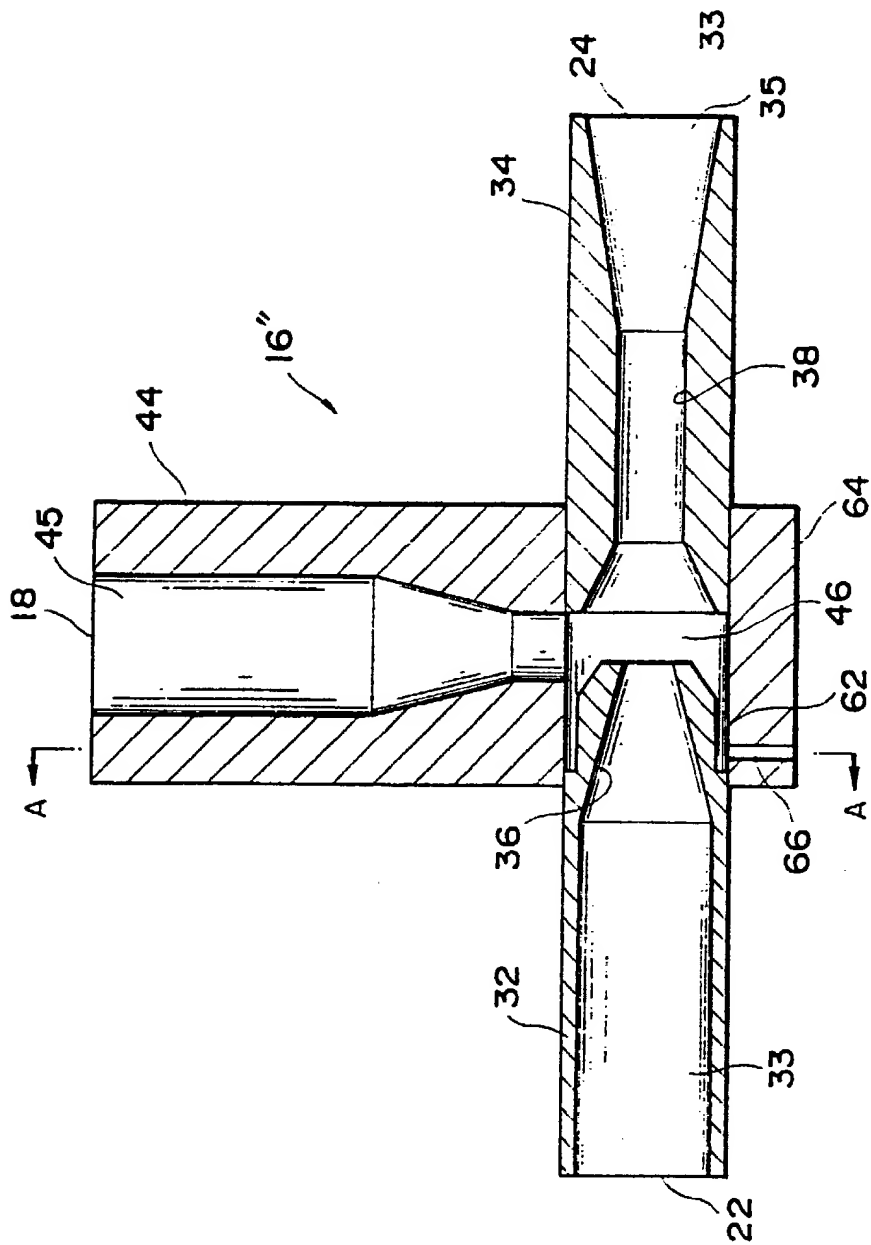
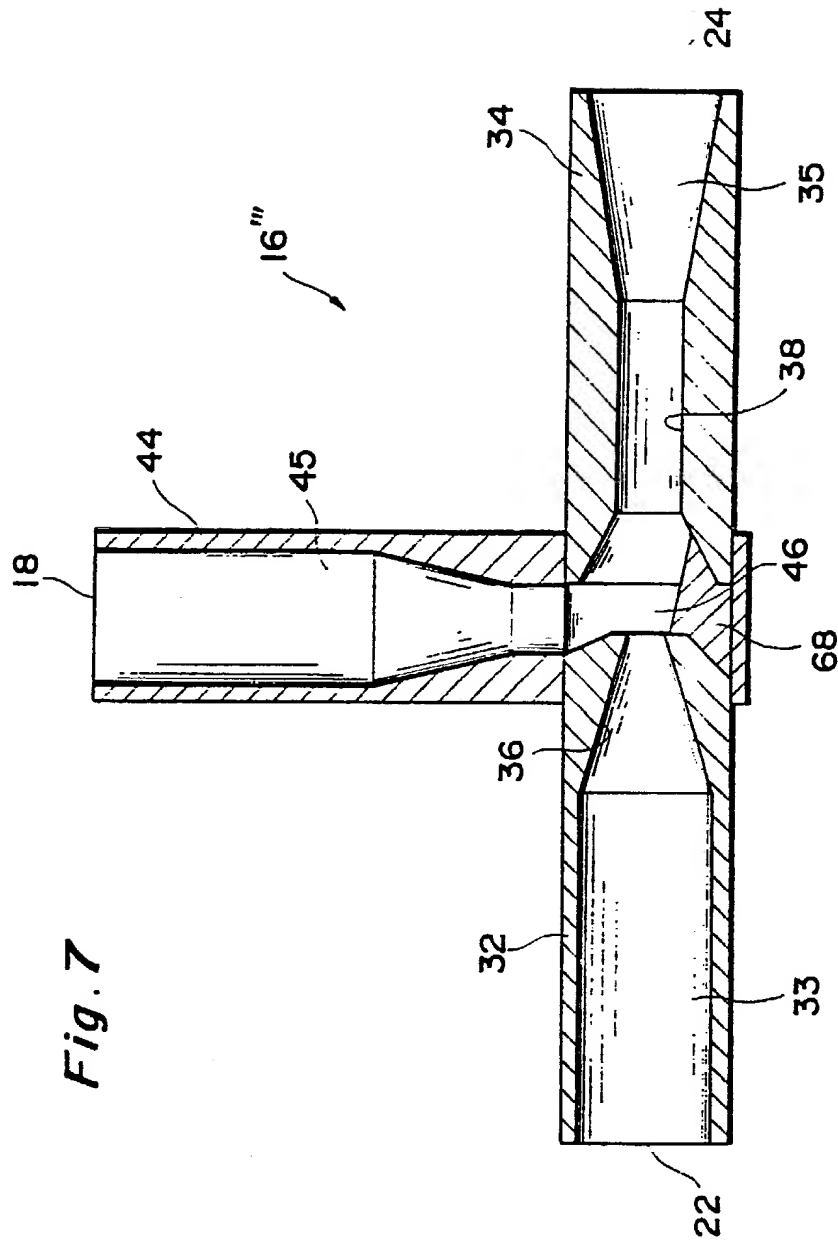
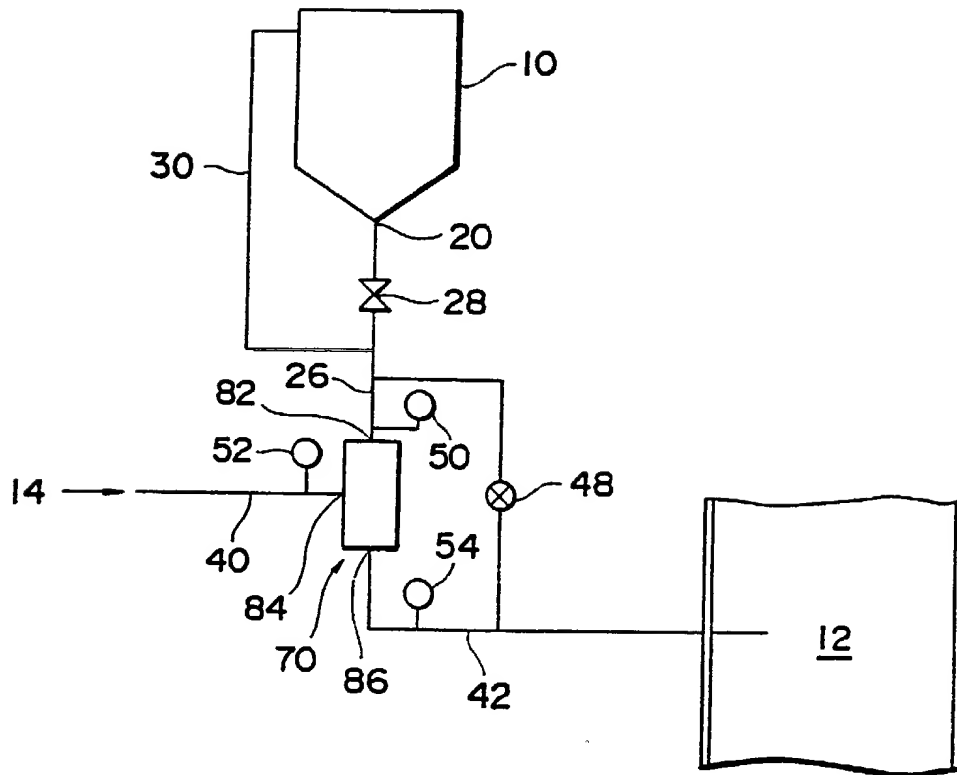


Fig. 5

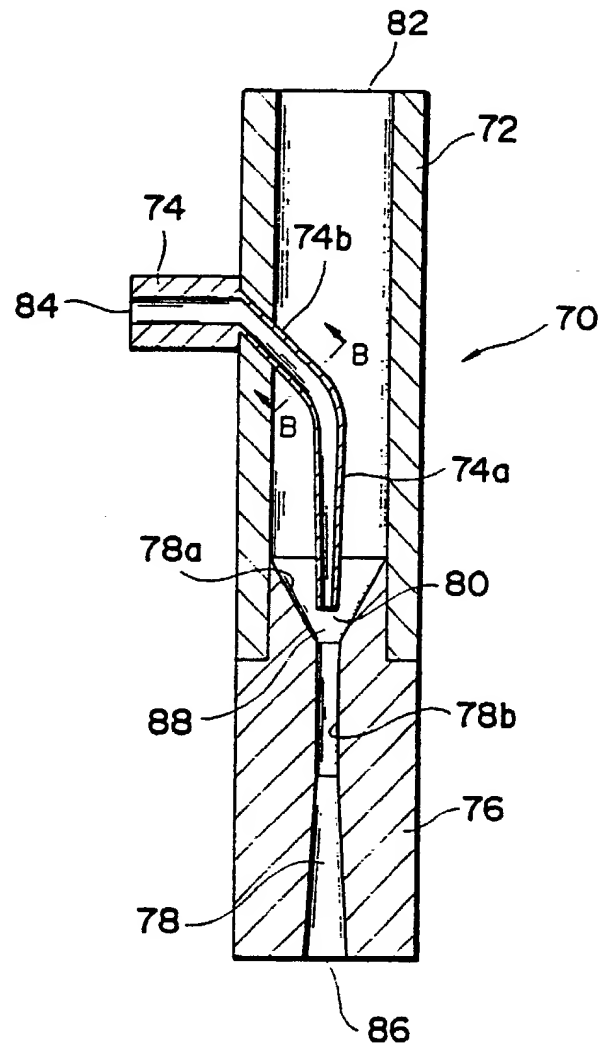




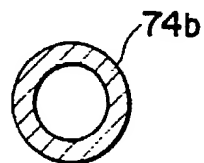
*Fig. 8*

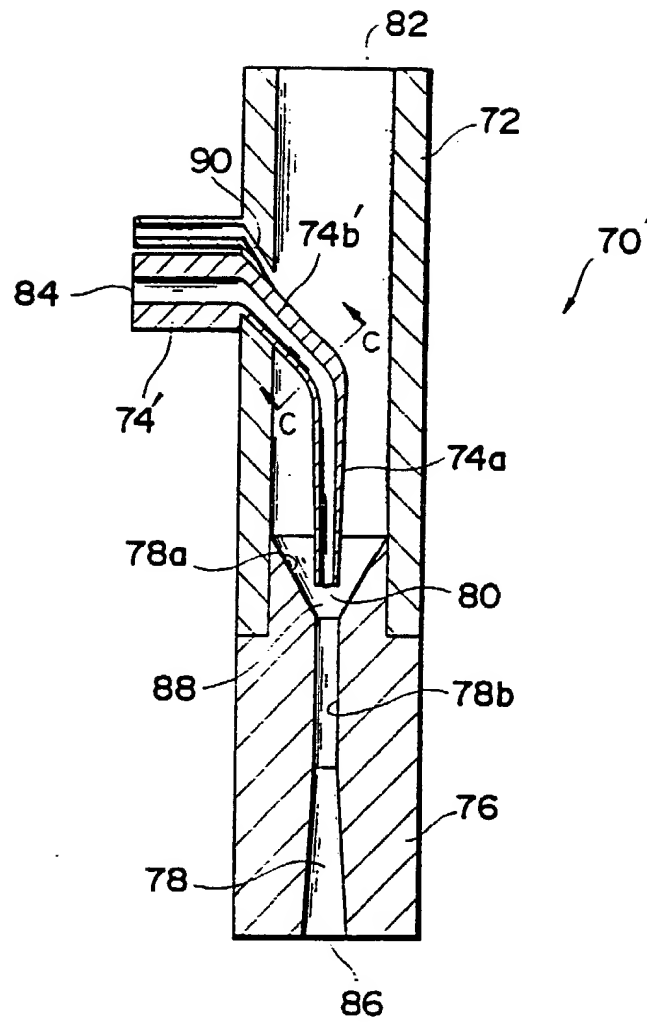
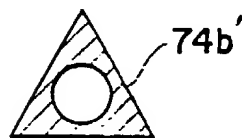


**Fig. 9**



**Fig. 10**



*Fig. 11**Fig. 12*

POWDER SUPPLY

The present invention relates to a powder catalyst supply system and a powder supply apparatus and, more particularly, to a powder catalyst supply system and a powder supply apparatus suitable for gas phase polymerization reaction of olefins, e.g., ethylene and propylene.

Related Background Art

In recent years, improvements in catalysts of transition metal for use in the polymerization of olefins have remarkably improved the productivity of olefin polymers per unit amount of the transition metal, consequently obviating the need for the removal of the catalyst after the polymerization.

When such highly active catalysts are used, olefins are generally polymerized in a gas phase since the polymerization reaction mixture can then be handled by the simplest procedure. Usually, fluidized bed reactors are widely used in the gas phase polymerization so as to effect the reaction smoothly. The olefin or an olefin-containing gas introduced into a lower portion of the reactor through a supply pipe is forced upward as uniformly distributed by a gas distributor plate to cause an olefin polymer and the catalyst in the form of polymer particles to mix within a fluidized bed for polymerization.

In the fluidized bed type reactor of this type, the powder catalyst must be continuously or intermittently supplied into the reactor under

pressure constantly by utilizing a powder catalyst supply system.

As is disclosed in Japanese Patent Publication No. 49-17426 and Japanese Patent Laid-Open  
5 No. 60-227824, in a conventional powder catalyst supply system, a powder catalyst is supplied under pressure from a powder catalyst storage container through a powder catalyst metering unit with an inert gas or a hydrogen gas, and the powder catalyst,  
10 together with a gas serving as the polymerization material, is supplied via an intermediate chamber and a Venturi tube.

Note that terms "polymerization" and "polymer" to be used herein include the meanings of  
15 "homopolymerization" and "copolymerization", and the meanings of "homopolymer" and "copolymer", respectively.

Since the conventional powder supply system has the arrangement as described above, the polymerization  
20 material gas can be gradually diffused into the pipe connected to the storage container and causes polymerization in this pipe or the powder catalyst storage container, so that an agglomerate can be sometimes undesirably formed.

#### 25 SUMMARY OF THE INVENTION

It is, therefore, a primary object of the present invention to provide a powder catalyst supply system and a powder supply apparatus which can prevent a material gas from being diffused or flowing backward

to the side of a catalyst storage container, thereby preventing the formation of a polymer agglomerate in a system line, and, what if a polymer agglomerate is generated, perceive the formation of a polymer agglomerate in a system line before that is exceeding,  
5 so supply a powder catalyst to a gas phase polymerization reactor efficiently.

According to the present invention, there is provided a powder catalyst supply system for supplying  
10 a powder catalyst to a gas phase polymerization reactor together with a gas serving as a polymerization material, comprising catalyst storage means for storing and supplying the powder catalyst, gas supply means for supplying the polymerization  
15 material gas, and a jet pump type powder supply apparatus for generating a negative pressure therein by accelerating a flow of the gas flowing from the gas supply means, drawing the powder catalyst from the catalyst storage means by the negative pressure, and  
20 supplying the powder catalyst to the gas phase polymerization reactor together with the gas.

With this arrangement, the powder catalyst is efficiently drawn by suction from the catalyst storage means and supplied to the gas phase polymerization reactor together with the material gas. Since the  
25 interior of the powder supply apparatus is set at the negative pressure, the material gas will not flow backward or be diffused from the powder supply apparatus to the catalyst storage means.

This system can employ a so-called ejector type powder supply apparatus comprising a catalyst suction flow path, connected to the catalyst storage means, for receiving the powder catalyst from the catalyst storage means, a gas feed flow path connected to the gas supply means, and a discharge flow path for discharging the drawn powder catalyst together with the gas sent from the gas supply means, wherein the gas feed flow path and the discharge flow path are coaxially arranged in series at a predetermined space therebetween, the gas feed flow path has a convergent nozzle formed at an end portion thereof opposing the discharge flow path, the discharge flow path has a diffuser formed at an end portion thereof opposing the nozzle of the gas feed flow path, and the catalyst suction flow path hydrodynamically communicates with the above space such that a central axis thereof intersects a common central axis of the gas feed flow path and the discharge flow path at a predetermined angle.

This system can also employ a so-called aspirator type powder supply apparatus in which a catalyst suction flow path and a discharge flow path are coaxially arranged in series, the discharge flow path has a diffuser formed at an end portion thereof opposing the catalyst suction flow path, and the gas feed flow path is formed in one gas feed pipe constituted by a first portion extending across the catalyst suction flow path and a second portion

integrally formed with the first portion and extending coaxially toward the discharge flow path in the catalyst suction flow path, the second portion of the gas feed pipe having a distal end formed with a convergent nozzle separated from the diffuser at a predetermined space.

These and other features and advantages of the present invention will become apparent to those skilled in the art upon a reading of the following detailed description when taken in conjunction with the drawings wherein there is shown and described an illustrative embodiment of the invention.

#### BRIEF DESCRIPTION OF THE DRAWINGS

In the course of the following detailed description, reference will be made to the attached drawings in which:

Fig. 1 is a view showing the schematic arrangement of a powder catalyst supply system using an ejector;

Fig. 2 is a sectional view showing the basic arrangement of an ejector according to the first embodiment of the present invention;

Fig. 3 is a sectional view showing the concreted arrangement of the ejector shown in Fig. 2;

Fig. 4 is a sectional view showing the basic arrangement of an ejector according to the second embodiment of the present invention;

Fig. 5 is a sectional view showing the basic arrangement of an ejector according to the third

embodiment of the present invention;

Fig. 6 is a sectional view taken along the line A - A of Fig. 5;

5 Fig. 7 is a sectional view showing the basic arrangement of an ejector according to the fourth embodiment of the present invention;

Fig. 8 is a view showing the schematic arrangement of a powder catalyst supply system using an aspirator;

10 Fig. 9 is a sectional view showing the basic arrangement of an aspirator according to the first embodiment of the present invention;

Fig. 10 is a sectional view taken along the line B - B of Fig. 9;

15 Fig. 11 is a sectional view showing the basic arrangement of an aspirator according to the second embodiment of the present invention; and

Fig. 12 is a sectional view taken along the line C - C of Fig. 11.

20 DESCRIPTION OF THE PREFERRED EMBODIMENTS

In the following description, like reference characters designate like or corresponding parts throughout the several views. Fig. 1 schematically shows the arrangement of a powder catalyst supply system according to the present invention which is  
25 used in a system for manufacturing a polymer, e.g., polyolefin. This powder catalyst supply system has a catalyst storage container or hopper 10 for storing a powder catalyst used for polymerization reaction,



e.g., the Ziegler-Natta type catalyst, a fluidized bed type gas phase polymerization reactor 12, and a gas supply source 14 for supplying a material gas, e.g., an olefin gas serving as the material for polymerization reaction. The gas supply source 14 and the gas phase polymerization reactor 12 are connected to each other through a jet pump type powder supply apparatus (to be described later), i.e., a so-called ejector 16, and a catalyst suction port 18 of the ejector 16 is connected to a catalyst outlet port 20 of the hopper 10. In brief, the ejector 16 generates, at its catalyst suction port 18, a negative pressure corresponding to the flow rate or the like of the material gas flowing from a gas feed port 22 to a discharge port 24. The catalyst drawn by suction through the catalyst suction port 18 is sent from the discharge port 24 into the gas phase polymerization reactor 12 together with the material gas.

The hopper 10 is provided with the catalyst outlet port 20 at its lowermost portion through which the powder catalyst is let to fall freely. The catalyst outlet port 20 and the catalyst suction port 18 of the ejector 16 are connected to each other through a first connecting pipe 26. The first connecting pipe 26 preferably has an inner diameter equal to 10 times or more, preferably 100 times the particle diameter of the powder catalyst so as to prevent the closing of the first connecting pipe 26 with the powder catalyst. The first connecting pipe

26 is connected such that the angle of its axis in the longitudinal direction with respect to the horizontal plane is equal to or larger than the angle of repose of the powder catalyst particle, and is more preferably 60° or more. In the most preferable case, as shown in Fig. 1, the first connecting pipe 26 is disposed such that the angle of its axis in the longitudinal direction with respect to the horizontal plane is 90°, i.e., is in the vertical direction.

The angle of repose is a maximum angle with respect to the horizontal plane, with which the surface layers of the group of powder particles can retain a stationary state by the friction with each other. More specifically, the angle of repose means an angle defined by the horizontal plane and the generating line or inclined surface of a circular cone which is formed when powder particles are let to continuously fall from a small hole or a gap to deposit on a flat surface (Takeshi Karino, "Powder Transportation Technique", Nikkan Kogyo Shinbun-sha, P.32).

As shown in Fig. 1, a valve 28 for shutting off communication of the powder catalyst is interposed in the first connecting pipe 26. In this embodiment, the powder catalyst is let to fall freely from the hopper 10. Therefore, a pressure equalization pipe 30 is connected between the hopper 10 and the first connecting pipe 26 in order to adjust the pressure difference between the hopper 10 and the first

connecting pipe 26.

Although not shown in Fig. 1, an inert gas or hydrogen gas is fed into the hopper 10 in order to prevent the powder catalyst from reacting in the hopper 10 and the first connecting pipe 26. This gas serves also as the carrier gas of the catalyst.

Fig. 2 shows the basic arrangement of the powder supplying ejector 16 which can be used in the powder catalyst supply system shown in Fig. 1. As is understood from Fig. 2, the ejector 16 has a gas feed pipe 32 whose interior serves as a feed flow path 33 of the material gas and a discharge pipe 34 whose interior serves as a discharge flow path 35 of the material gas and the powder catalyst. A convergent nozzle 36 is formed at the end portion of the gas feed pipe 32 on the discharge pipe 34 side, and a diffuser 38 is formed on the discharge pipe 34. The gas feed pipe 32 and the discharge pipe 34 are connected coaxially and in series, and the outlet portion of the nozzle 36 and the inlet portion of the diffuser 38 are opposed to each other at a predetermined gap. The end portion of the gas feed pipe 32 on the opposite side to the nozzle 36, i.e., the gas feed port 22, is connected to a second connecting pipe 40 extending from the gas supply source 14. The discharge port 24 of the discharge pipe 34 is connected to the gas phase polymerization reactor 12 through a third connecting pipe 42.

The ejector 16 also has a catalyst suction pipe

44 to be connected to the first connecting pipe 26. The interior of the catalyst suction pipe 44 serves as a suction flow path 45 of the powder catalyst, and is in fluid communication with a space defined between the gas feed pipe 32 and the discharge pipe 34, i.e., a so-called catalyst suction chamber 46. The central axis of the catalyst suction pipe 44 is perpendicular to the common central axis of the gas feed pipe 32 and the discharge pipe 34. Accordingly, this ejector 16 has a T-shape.

For use, this ejector 16 having the T-shape is arranged such that the central axis of the catalyst suction pipe 44 is coaxial with the central axis of the first connecting pipe 26. Accordingly, the catalyst suction port 18 of the catalyst suction pipe 44 is set upward in the vertical direction, and the common axis of the gas feed pipe 32 and the discharge pipe 34 is set in the horizontal direction.

With this arrangement, even when no external force is applied, the powder catalyst flows from the hopper 10 to freely fall into the catalyst suction chamber 46 of the ejector 16 through the first connecting pipe 26 and the catalyst suction pipe 44 of the ejector 16. In order to prevent the fallen catalyst from being deposited to retain on the end portion of the gas feed pipe 32 on the side of nozzle 36, the end face of the gas feed pipe 32 on the nozzle 36 side is arranged as close as possible to the gas supply source 14 (on the left side in Fig. 2). It is

effective to locate the end face of the gas feed pipe 32 at a position at least not exceeding the central axis of the catalyst suction pipe 44. For the same purpose, it is preferable to set the gas feed pipe 32 to have a tapered conical outer shape at its end portion on the nozzle 36 side.

The powder catalyst flows through the catalyst suction pipe 44 and the discharge pipe 34. Therefore, in order to prevent the interiors of the catalyst suction pipe 44 and the discharge pipe 34 from clogging with the powder catalyst, the minimum inner diameters of the catalyst suction pipe 44 and the discharge pipe 34 are preferably set to 10 times or more, preferably 100 times the particle diameter of the powder catalyst, in the same manner as in the first connecting pipe 26.

As shown in Fig 1, a differential pressure gauge 48 is provided between the first and third connecting pipes 26 and 42 to measure the pressure difference between the discharge port 24 and the catalyst suction port 18 of the ejector 16. Furthermore, pressure gauges 50, 52, and 54 are provided respectively to the first, second, and third connecting pipes 26, 40, and 42 to measure the respective pressures at the catalyst suction port 18, the gas feed port 22, and the discharge port 24 of the ejector 16, respectively.

In the powder catalyst supply system having the arrangement as described above, while the shut-off valve 28 in the first connecting pipe 26 is open, when

the material gas is supplied from the gas supply source 14 to the gas phase polymerization reactor 12, the powder catalyst flows from the hopper 10 into the ejector 16 through the first connecting pipe 26 by the gravity and is at the same time drawn into the ejector 16 by the suction of the ejector 16. More specifically, when the material gas flows into the gas feed pipe 32 of the ejector 16 and is injected from the nozzle 36, its flow velocity is increased in the catalyst suction chamber 46, and the pressure in the catalyst suction chamber 46 is decreased to be lower than that in the hopper 10. As a result, the powder catalyst can be reliably drawn by suction from the hopper 10 into the catalyst suction chamber 46 of the ejector 16. Since the pressure in the catalyst suction chamber 46 is lower than that in the hopper 10, the material gas will not be diffused toward the hopper 10. The powder catalyst drawn into the catalyst suction chamber 46 is entrained in the material gas injected from the nozzle 36 and is fed to the discharge pipe 34. The flow velocity of this material gas containing the catalyst is decreased by the diffuser 38, and the pressure thereof is increased. Thereafter, the material gas containing the catalyst is sent into the gas phase polymerization reactor 12 through the third connecting pipe 42.

When the powder catalyst is drawn into the catalyst suction chamber 46, since the distal end of the gas feed pipe 32 is arranged at positions as

remote as possible from the central axis of the catalyst suction pipe 44, as shown in Fig. 2, the powder catalyst is suppressed from being deposited on the outer surface of the nozzle 36. Accordingly, most  
5 of the catalyst sent from the hopper 10 is sent to the gas phase polymerization reactor 12.

At this time, if no abnormality occurs, the pressure balance in the powder catalyst supply system satisfies

10 
$$P_2 > P_3 > P_1$$

where  $P_1$  is the pressure at the catalyst suction port 18 of the ejector 16,  $P_2$  is the pressure at the gas feed port 22,  $P_3$  is the pressure at the discharge port 24. In this case, a pressure difference  $\Delta P$  between  
15 the pressure  $P_1$  at the catalyst suction port 18 and the pressure  $P_3$  at the discharge port 24 is determined by the mass and flow rate of the material gas, the size and shape of the nozzle 36 of the gas feed pipe 32, the size and shape of the diffuser 38 of the  
20 discharge pipe 34, and the like. Normally, since the pressure  $P_3$  at the discharge port 24 is determined depending on the pressure in the gas phase polymerization reactor 12, the pressure  $P_1$  at the catalyst suction port 18 can be determined  
25 accordingly.

These pressures  $P_1$ ,  $P_2$ , and  $P_3$ , and the pressure difference  $\Delta P$  can be constantly monitored by the pressure gauges 50, 52, and 54, and the differential pressure gauge 48 described above, and an abnormality

in a pipe can be detected from a variation in these pressures. For example, when the material gas is supplied from the gas supply source 14 at a predetermined flow rate, if P2 and/or P3 is increased, generation of an agglomerate in the line between the gas supply source 14 and the gas phase polymerization reactor 12 can be detected. Then, troubles, e.g., clogging of a pipe, can be prevented.

When the pressure difference  $\Delta P$  is decreased due to a cause such that the third connecting pipe 42 extending into the gas phase polymerization reactor 12 clogs, the material gas can undesirably flow backward from the catalyst suction chamber 46 of the ejector 16 to the first connecting pipe 26. Hence, whether or not the reverse flow of the material gas can occur can be determined by constantly monitoring the pressure difference  $\Delta P$  by the differential pressure gauge 48. If the material gas should possibly flow backward to the hopper 10, the shut-off valve 28 may be closed immediately.

Fig. 2 shows the basic arrangement of the ejector 16. An example of a detailed concrete arrangement of the ejector 16 is shown in Fig. 3. An ejector 16 shown in Fig. 3 has, in addition to a catalyst suction pipe 44, a gas feed pipe 32 and a discharge pipe 34 similar to those described above, a body block 56. A T-shaped through hole 58 is formed in the body block 56. Portions 58a and 58b of the through hole 58 are coaxially arranged, and a portion 58c is perpendicular



to the portions 58a and 58b. Threaded portions 59a, 59b, and 59c are formed on the portions 58a, 58b, and 58c, respectively. Threaded portions 60a, 60b, and 60c formed on the outer circumferential surfaces of the gas feed pipe 32, discharge pipe 34 and catalyst suction pipe 44 are threadably engaged with the threaded portions 59a, 59b, and 59c, respectively. Thus, the gas feed pipe 32, the discharge pipe 34 and the catalyst suction pipe 44 are fixed to the body block 56 in the same order as in Fig. 2. Threaded portions 61a, 61b, and 61c are preferably formed on the outer circumferential surfaces of the exposed end portions of the gas feed pipe 32, the discharge pipe 34, and the catalyst suction pipe 44, respectively. Although not shown, these threaded portions 61a, 61b, and 61c are used to couple the first, second and third first connecting pipes 26, 40, and 42 to the ejector 16 by using nipple joints or the like.

The arrangement of the ejector 16 shown in Fig. 3 is illustrative and not restrictive. Therefore, an ejector 16 may be integrally formed with a catalyst suction pipe 44, a gas feed pipe 32, and a discharge pipe 34. If the ejector is integrally formed with these pipes, leakage of the gas and the catalyst can be completely prevented.

Fig. 4 shows the basic arrangement of an ejector according to the second embodiment of the present invention. This ejector 16' is substantially the same as the ejector 16 shown in Fig. 2 except that a

catalyst suction pipe 44 is inclined to the left side of Fig. 4. The angle defined by the central axes of the catalyst suction pipe 44 and a gas feed pipe 32 is 30° or more, and is preferably equal to or larger than the angle of repose of the powder catalyst particles. The operation of the ejector 16' is the same as the ejector 16 shown in Fig. 2, and a catalyst transported through the catalyst suction pipe 44 is fed into a discharge pipe 34 comparatively smoothly. Although the ejector 16 of Fig. 2 has advantages such that the manufacture and piping plan of the system are facilitated, an energy loss occurs since the traveling direction of the powder catalyst is changed by about 90° in the catalyst suction chamber 46. In contrast to this, in the ejector 16' of Fig. 4, accumulation of the catalyst on the end portion of the gas feed pipe 32 on the nozzle 36 side is suppressed due to the smooth flow of the catalyst.

Figs. 5 and 6 show an ejector according to the third embodiment of the present invention. As is understood from Figs. 5 and 6, this ejector 16" is substantially the same as the ejector 16 shown in Fig. 2 except that the outer diameter of the end portion of a gas feed pipe 32 on a nozzle 36 side is decreased and that an annular space 62 is formed around it. This space 62 is in fluid communication with a hole 66 formed in a surrounding portion 64. The surrounding portion 64 defines a catalyst suction chamber 46 by surrounding the opposing end portions of

the gas feed pipe 32 and a discharge pipe 34. A small amount of suitable gas, e.g., material gas, inert gas or hydrogen gas, is fed from a gas supply source (not shown) into the hole 66.

5           When a small amount of material gas, inert gas or hydrogen gas is continuously or intermittently fed into the hole 66, this gas is injected into the catalyst suction chamber 46 through the space 62. The catalyst retaining on the circumferential portion of  
10           the end portion of the gas feed pipe 32 can be removed by this injected flow. The removed catalyst is fed from the catalyst suction chamber 46 into the discharge pipe 34 by the flow of the material gas flowing from the gas feed pipe 32.

15           The gas for removing the catalyst is preferably an inert gas as it can undesirably flow backward to a catalyst suction pipe 44. In this case, an inexpensive dry nitrogen gas is desirable. The supply amount, the supply pressure, and the like of the gas  
20           for removing the catalyst must be determined such that the pressure in the catalyst suction chamber 46 will not be increased to flow the material gas in the catalyst suction chamber 46 to the hopper 10.

25           Fig. 7 shows an ejector according to the fourth embodiment of the present invention. In this ejector 16", a retention preventive member 68 for preventing retention of the catalyst is provided to a portion in a catalyst suction chamber 46 opposite to a catalyst suction pipe 44. The retention preventive member 68

fills the space extending from a portion under the end portion of a gas feed pipe 32 on a nozzle 36 side to the inlet portion of a discharge pipe 34. The upper surface of the retention preventive member 68 is inclined downward in the flow direction of the material gas, and a portion thereof adjacent to the gas feed pipe 32 is at the same level with the position of the edge portion of the outlet of the nozzle 36 on the lowermost side.

In an ejector not having a retention preventive member 68, i.e., an ejector as shown in Fig. 2, a small amount of powder catalyst tends to retain under the end portion of the gas feed pipe 32 on the nozzle 36 side. In contrast to this, in the ejector 16" having the retention preventive member 68, the space under the nozzle 36 is eliminated by the retention preventive member 68, so that the catalyst is prevented from entering this portion. The powder catalyst is smoothly guided to the discharge pipe 34 along the inclined upper surface of the retention preventive member 68. In this embodiment, the upper surface of the retention preventive member 68 is a flat surface. However, it will be understood that the upper surface of the retention preventive member 68 can have other shapes, e.g., a recessed surface.

An experiment will be described wherein a powder catalyst supply system according to the present invention is used in a system for manufacturing linear low-density polyethylene by gas phase copolymerization

of ethylene and 1-butene. In this experiment, the powder catalyst supply system is of the type as shown in Fig. 1, and an ejector 16' used in this powder catalyst supply system is of the type as shown in Fig. 4. Note that this ejector 16' is provided with a gas feed hole for removing the powder catalyst accumulated on the upper surface of the end portion of a gas feed pipe 32 on the nozzle 36 side. A dry nitrogen gas was fed into this gas feed hole and constantly injected into a catalyst suction chamber 46 at an injection amount of 1.0 Nm<sup>3</sup>/hr.

The inner diameter of a catalyst suction port 18 of the ejector 16 was set to 25 mm, the minimum inner diameter of the nozzle 36 of the gas feed pipe 32 was set to 10 mm, and the minimum inner diameter of a diffuser 38 of a discharge pipe 34 was set to 15 mm. The angle defined by the axis of a catalyst suction pipe 44 and the axis of the gas feed pipe 32 was set to 45°. The material gas was a gas mixture of ethylene and 1-butene, and the catalyst was the Ziegler-Natta type catalyst.

The material gas was supplied from a gas supply source 14 at a mass flow rate of 1000 kg/hr when a pressure P2 at a gas feed port 22 and a pressure P3 at a discharge port 24 were 25 kgf/cm<sup>2</sup>G and 20 kgf/cm<sup>2</sup>G, respectively. Then, a pressure difference  $\Delta P$  between the pressure P1 at the catalyst suction port 18 and the pressure P3 at the discharge port 24 was 1.0 kgf/cm<sup>2</sup>. In this case, the pressure P1 was

19 kgf/cm<sup>2</sup>G.

When the polyethylene manufacturing system was operated under these conditions, stable polyethylene was manufactured over a long period of time without forming an agglomerate of a polymer in the ejector 16 and in a first connecting pipe 26 coupling the ejector 16 and a hopper 10.

The powder catalyst supply system can employ a jet pump type powder supply apparatus other than an ejector. Fig. 8 schematically shows a powder catalyst supply system using a so-called aspirator (to be described in detail hereinbelow) as the powder supply apparatus. This powder catalyst supply system is substantially the same as the system shown in Fig. 1 except that the ejector is replaced with the aspirator. Therefore, in this powder catalyst supply system, the same or corresponding portions as in the system shown in Fig. 1 are denoted by the same reference numerals, and a detailed description thereof will be omitted.

Fig. 9 shows the basic arrangement of a preferred embodiment of the aspirator used in the powder catalyst supply system of Fig. 8. This aspirator 70 has a catalyst suction pipe 72 connected to a first connecting pipe 26 extending from a hopper 10, a gas feed pipe 74 connected to a second connecting pipe 40 extending from a gas supply source 14, and a discharge pipe 76 connected to a third connecting pipe 42 extending from a gas phase polymerization reactor 12.

The catalyst suction pipe 72 and the discharge pipe 76 are coaxially arranged in series and are integrally coupled to each other. The inner wall surface of the catalyst suction pipe 72 is preferably straight and cylindrical in order to smoothly flow the catalyst. The discharge pipe 76 is provided with a diffuser 78 for converting the kinetic energy of the fluid flowing from the catalyst suction pipe 72 to a pressure energy. The diffuser 78 is coaxially formed with the catalyst suction pipe 72.

An inlet portion 78a of the diffuser 78 has a conical recessed surface. When the catalyst suction pipe 72 and the discharge pipe 76 are set in the vertical direction, the angle of the generating line of the inner wall surface of the inlet portion 78a with respect to the horizontal plane is set equal to or larger than the angle of repose of the powder catalyst particles, and is more preferably 60° or more. Then, the powder catalyst freely falling through the catalyst suction pipe 72 is smoothly guided from the conical inlet portion 78a to a throat portion 78b of the diffuser 78.

The gas feed pipe 74 extends into the catalyst suction pipe 72 through the appropriate portion of the pipe wall of the catalyst suction pipe 72. The gas feed pipe 74 is hermetically fixed to the catalyst suction pipe 72 at the portion where it extends through the catalyst suction pipe 72. A convergent nozzle 80 is formed at the end portion of the gas feed

pipe 74 inside the catalyst suction pipe 72. This nozzle 80 is directed to the diffuser 78 of the discharge pipe 76. The axis of a portion 74a of the gas feed pipe 74 having the nozzle 80 is coaxial with  
5 the catalyst suction pipe 72 and the diffuser 78 of the discharge pipe 76. The nozzle 80 is arranged to oppose the throat portion 78b of the diffuser 78 at a predetermined gap.

As shown in Fig. 10, the gas feed pipe 74  
10 typically has a circular section. A portion 74b of the gas feed pipe 74 between the pipe wall of the catalyst suction pipe 72 and the portion 74a of the gas feed pipe 74 is inclined toward the diffuser 78 (to the lower side in Fig. 9). It is effective that  
15 the angle of the inclined portion 74b with respect to the horizontal plane is equal to or larger than the angle of repose of the powder catalyst when the catalyst suction pipe 72 is set in the vertical direction. Hence, the catalyst naturally falling from  
20 the catalyst suction pipe 72 smoothly slides down to the diffuser 78 without being deposited on the gas feed pipe 74 even if no external force is applied.

The minimum size of the gap between the nozzle 80 of the gas feed pipe 74 and the inner circumferential  
25 surface of the catalyst suction pipe 72 around the nozzle 80, or between the nozzle 80 and the conical recessed surface 78a of the discharge pipe 76 is set to 10 times or more, preferably 100 times the particle diameter of the powder catalyst to prevent the pipes



from clogging with the powder catalyst. For the same purpose, the minimum inner diameter of the throat portion 78b of the diffuser 78 of the discharge pipe 76 is also set to 10 times or more, preferably 100 times the particle diameter of the powder catalyst.

When the aspirator 70 having the arrangement as described above is to be used, a suction port 82 of its catalyst suction pipe 72 is coaxially connected to the first connecting pipe 26. Since the first connecting pipe 26 is normally set in the vertical direction, the catalyst suction pipe 72 and the discharge pipe 76 are also set in the vertical direction. The horizontal second connecting pipe 40 is connected to a gas feed port 84 of the gas feed pipe 74. Furthermore, the third connecting pipe 42 is connected to a discharge port 86 at the lower end of the discharge pipe 76.

In operation of the powder catalyst supply system having the aspirator 70 described above, when the material gas is supplied from the gas supply source 14 to the gas phase polymerization reactor 12 while a shut-off valve 28 of the first connecting pipe 26 is open, the powder catalyst falls from the hopper 10 into the aspirator 70 through the first connecting pipe 26 and is simultaneously drawn into the aspirator 70 by the suction operation of the aspirator 70. In the same manner as in the ejector described above, when the material gas is injected from the nozzle 80 of the gas feed pipe 74 to the diffuser 78 of the

discharge pipe 76, the space between the nozzle 80 and the diffuser 78, i.e., a catalyst suction chamber 88 is set at a negative pressure, so that the powder catalyst can be drawn. The powder catalyst guided  
5 into the catalyst suction chamber 88 is accompanied with the material gas injected from the nozzle 80, is guided into the discharge pipe 76, and is sent to the gas phase polymerization reactor 12 through the third connecting pipe 42. The flow velocity of the material  
10 gas containing the catalyst is decreased by the diffuser 78 of the discharge pipe 76, so that the pressure of the material gas is increased.

Since the aspirator 70 is arranged in the above manner considering that the material to be dealt with  
15 may contain not only a gas but also a powder, the powder catalyst will hardly retain in the aspirator 70. Since the pressure at the suction port 82 of the aspirator 70 is lower than that in the hopper 10, the material gas will not be undesirably diffused to the  
20 hopper 10.

In the powder catalyst supply system using this aspirator 70 as well, it is preferable to provide pressure gauges 50, 52, and 54 for measuring the  
25 respective pressures at the suction port 82 of the aspirator 70, the gas feed port 84, and the discharge port 86, and a differential pressure gauge 48 for measuring the pressure difference between the suction port 82 and the discharge port 86 in the same manner as in the system shown in Fig. 1, so that troubles,

e.g., clogging of the catalyst or an agglomerate of a polymer in the pipes, can be prevented. The pressure gauges 50, 52, and 54, and the differential pressure gauge 48 are used in the same manner as described above.

Fig. 11 shows another embodiment of an aspirator. In this aspirator 70', as shown in Fig. 12, a portion 74b' of a gas feed pipe 74' located in a catalyst suction pipe 72 has a triangular cross-section. One of the sides of the portion 74b' of the gas feed pipe 74', which has the shape of a triangular prism, is directed to the catalyst suction port 82, i.e., to the upper side of Fig. 11.

In the gas feed pipe 74' having this shape, it will be readily understood that the amount of powder catalyst accumulated on the gas feed pipe 74' can be greatly decreased to be smaller than in a gas feed pipe having a circular section. This gas feed pipe can have any sectional shape as far as a powder cannot be easily deposited in it, and may have, e.g., a rhombic or elliptic shape.

A through hole 90 is formed in a portion of the pipe wall of the catalyst suction pipe 72 above the gas feed pipe 74'. A supply source (not shown) of an appropriate gas, e.g., the material gas, the inert gas, or the hydrogen gas, is connected to this through hole 90. A small amount of material gas or the like is injected from the outlet of the through hole 90 toward the inclined portion 74b' of the gas feed pipe

74'. Hence, the powder catalyst deposited on the gas feed pipe 74', especially on the inclined portion 74b' can be blown off to be guided into a discharge pipe 76.

5           The gas to be used is preferably an inexpensive dry nitrogen gas. The amount of gas to be fed should be determined such that the pressure in a catalyst suction chamber 88 will not be excessively increased.

10           Excluding the above respects, the aspirator 70' is substantially the same as the aspirator shown in Fig. 9.

15           An experiment will be described wherein a powder catalyst supply system having an aspirator is used in a system for manufacturing linear low-density polyethylene by gas phase copolymerization of ethylene and 1-butene. In this experiment, the powder catalyst supply system is of the type as shown in Fig. 8, and an aspirator 70 used in this powder catalyst supply system is of the type as shown in Fig. 9.

20           The inner diameter of a catalyst suction port 82 of the aspirator 70 was set to 60 mm, the outer and inner diameters of a nozzle 80 of a gas feed pipe 74 were set to 15 mm and 10 mm, respectively, and the minimum inner diameter of a diffuser 78 of a discharge pipe 76 was set to 12 mm. The angle defined by the axis of a catalyst suction pipe 72 and the axis of an inclined portion 74b of the gas feed pipe 74 was set to 40°, and the angle of the generating line of the conical surface of an inlet portion 78a of the

25

diffuser 78 with respect to the horizontal direction was set to 60°. The material gas was a gas mixture of ethylene and 1-butene, and the catalyst was the Ziegler-Natta type catalyst.

5           The material gas was supplied from a gas supply source 14 at a mass flow rate of 1000 kg/hr when a pressure P2 at a gas feed port 84 and a pressure P3 at a discharge port 86 were 25 kgf/cm<sup>2</sup>G and 20 kgf/cm<sup>2</sup>G, respectively. Then, a pressure difference  $\Delta P$  between  
10           a pressure P1 at a catalyst suction port 82 and the pressure P3 at the discharge port 86 was 1.0 kgf/cm<sup>2</sup>. In this case, the pressure P1 was 19 kgf/cm<sup>2</sup>G.

          When the polyethylene manufacturing system was operated under these conditions, stable polyethylene  
15           was manufactured over a long period of time without forming an agglomerate of a polymer in the aspirator 70 and in a first connecting pipe 26 coupling the aspirator 70 and a hopper 10.

          In the above embodiments, the powder catalyst is  
20           directly sent from the hopper 10 to an ejector 16 or the aspirator 70. However, a catalyst metering unit, e.g., a metering pipe, may be interposed between the hopper 10 and the ejector 16 or the aspirator 70. The catalyst may be supplied to the ejector 16 or the  
25           aspirator 70 not by gravity-drop, but by using an appropriate gas under pressure, e.g., an inert gas or the hydrogen gas. In the latter case, no pressure equalization pipe 30 is needed.

          Furthermore, it will be apparent to those skilled

in the art that the ejector or aspirator described above can be used not only in the powder catalyst supply system but also in other powder transportation systems.

5           From the invention thus described, it will be obvious that the invention may be varied in many ways. Such variations are not to be regarded as a departure from the spirit and scope of the invention, and all  
10 such modifications as would be obvious to one skilled in the art are intended to be included within the scope of the following claims.

WHAT IS CLAIMED IS:

1. A powder catalyst supply system for supplying  
a powder catalyst to a gas phase polymerization  
reactor together with a gas serving as a  
5 polymerization material, comprising:

catalyst storage means for storing and supplying  
the powder catalyst;

gas supply means for supplying the polymerization  
material gas; and

10 a jet pump type powder supply apparatus for  
generating a negative pressure therein by accelerating  
a flow of the gas flowing from said gas supply means,  
drawing the powder catalyst from said catalyst storage  
means by the negative pressure, and supplying the  
15 powder catalyst to said gas phase polymerization  
reactor together with the gas.

2. A system according to claim 1, wherein said  
powder supply apparatus comprises:

a catalyst suction flow path connected to said  
20 catalyst storage means through a first pipe;

a gas feed flow path connected to said gas supply  
means through a second pipe; and

a discharge flow path connected to said gas phase  
polymerization reactor through a third pipe,

25 wherein said gas feed flow path and said  
discharge flow path are coaxially arranged in series  
at a predetermined space therebetween,

said gas feed flow path has a convergent nozzle  
formed at an end portion thereof opposing said

discharge flow path,

said discharge flow path has a diffuser formed at an end portion thereof opposing said nozzle of said gas feed flow path, and

5        said catalyst suction flow path being in fluid communication with said space such that a central axis thereof intersects a common central axis of said gas feed flow path and said discharge flow path at a predetermined angle.

10        3. A system according to claim 2, wherein the angle defined by the common central axis of said gas feed flow path and said discharge flow path and the central axis of said catalyst suction flow path is 90°.

15        4. A system according to claim 2, wherein the angle defined by the central axis of said gas feed flow path and the central axis of said catalyst suction flow path is less than 90°.

20        5. A system according to any one of claims 2 to 4, wherein said nozzle is arranged such that a distal end thereof is retreated from the central axis of said catalyst suction flow path in a direction to be separated from said discharge flow path.

25        6. A system according to any one of claims 2 to 5, wherein minimum inner diameters of said catalyst suction flow path and said discharge flow path are not less than 10 times a powder particle diameter of the powder catalyst.

7. A system according to any one of claims 2 to



6, wherein said powder supply apparatus is arranged such that an inlet of said catalyst suction flow path is directed upward.

5        8. A system according to claim 7, wherein an angle defined by the central axis of said catalyst suction flow path with respect to a horizontal plane is set to not less than an angle of repose of the powder catalyst.

10       9. A system according to claim 7, wherein an angle defined by the central axis of said catalyst suction flow path with respect to a horizontal plane is set to not less than 60°.

10. A system according to any one of claims 2 to 9, further comprising:

15       gas injecting means, provided to said powder supply apparatus, for injecting a catalyst removing gas to remove the powder catalyst retaining around said nozzle; and

20       gas supply means for supplying the catalyst removing gas to said gas injecting means.

11. A system according to claim 10, wherein the catalyst removing gas is a gas serving as a polymerization material.

25       12. A system according to claim 10, wherein the catalyst removing gas is an inert gas or a hydrogen gas.

13. A system according to claim 12, wherein the inert gas is dry nitrogen gas.

14. A system according to any one of claims 2 to

13, wherein said powder supply apparatus is provided with a retention preventive member for preventing the powder catalyst from retaining at a lower portion of a gap defined around said nozzle.

5           15. A system according to claim 14, wherein said retention preventive member extends from said lower portion of the gap defined around said nozzle to an inlet portion of said diffuser through a lower portion of the space between said gas feed flow path and said  
10           discharge flow path.

16. A system according to claim 1, wherein said powder supply apparatus comprises:

          a catalyst suction flow path connected to said catalyst storage means through a first pipe;

15           a gas feed flow path connected to said gas supply means through a second pipe; and

          a discharge flow path connected to said gas phase polymerization reactor through a third pipe,

          wherein said catalyst suction flow path and said  
20           discharge flow path are coaxially arranged in series,

          said discharge flow path has a diffuser formed at an end portion thereof opposing said catalyst suction flow path, and

          said gas feed flow path is formed in one gas feed  
25           pipe constituted by a first portion extending across said catalyst suction flow path and a second portion integrally formed with said first portion and extending in said catalyst suction flow path coaxially to said discharge flow path, said second portion of

said gas feed pipe having a distal end formed with a convergent nozzle separated from said diffuser at a predetermined space.

5        17. A system according to claim 16, wherein said catalyst suction flow path is substantially straight and cylindrical.

10       18. A system according to one of claims 16 and 17, wherein a minimum size of a portion around said second portion of said gas feed flow path and a minimum inner diameter of said discharge flow path are set to not less than 10 times the powder particle diameter of the powder catalyst.

15       19. A system according to any one of claims 16 to 18, wherein said powder supply apparatus is arranged such that an inlet of said catalyst suction flow path is directed upward and a central axis of said catalyst suction flow path is set in the vertical direction.

20       20. A system according to claim 19, wherein said first portion of said gas feed pipe is inclined downward so that an angle thereof with respect to a horizontal plane is not less than an angle of repose of the powder catalyst.

25       21. A system according to any one of claims 16 to 20, wherein said first portion of said gas feed pipe has a circular cross-section.

22. A system according to any one of claims 16 to 20, wherein said first portion of said gas feed pipe has a triangular cross-section, and said gas feed pipe is arranged such that a vertex of the triangular

cross-section is directed to an inlet of said catalyst suction flow path.

23. A system according to any one of claims 16 to 22, further comprising:

5        gas injecting means, provided to said powder supply apparatus, for injecting a catalyst removing gas to remove the powder catalyst deposited on said gas feed pipe; and

10        gas supply means for supplying the catalyst removing gas to said gas injecting means.

24. A system according to claim 23, wherein the catalyst removing gas is a gas serving as a polymerization material.

15        25. A system according to claim 23, wherein the catalyst removing gas is an inert gas or a hydrogen gas.

26. A system according to claim 25, wherein the inert gas is dry nitrogen gas.

20        27. A system according to one of claims 19 and 20, wherein an inlet portion of said discharge flow path has a conical recessed surface, and an angle of a generating line of said recessed surface with respect to a horizontal plane is not less than an angle of repose of the powder catalyst.

25        28. A system according to one of claims 19 and 20, wherein an inlet portion of said discharge flow path has a conical recessed surface, and an angle of a generating line of said recessed surface with respect to a horizontal plane is not less than 60°.

29. A system according to any one of claims 2 to 28, comprising pressure gauges for respectively detecting a pressure in said catalyst suction flow path, a pressure in said gas feed flow path, and a pressure in said discharge flow path.

30. A system according to any one of claims 2 to 29, comprising a differential pressure gauge for detecting a pressure difference between the pressure in said catalyst suction flow path and the pressure in said discharge flow path.

31. A system according to any one of claims 1 to 30, wherein the powder catalyst is drawn by said powder supply apparatus from said catalyst storage means together with an inert gas or nitrogen gas.

32. A powder supply apparatus for generating a negative pressure therein by accelerating a flow of a gas, drawing a powder by the negative pressure, and discharging the powder together with the gas, comprising:

a powder suction flow path adapted to be connected to powder storage means to receiving the powder from said powder storage means;

a gas feed flow path adapted to be connected to gas supply means; and

a discharge flow path for discharging the drawn powder together with the gas sent from said gas supply means,

wherein said powder suction flow path and said

discharge flow path are coaxially arranged in series,  
said discharge flow path has a diffuser formed at  
an end portion thereof opposing said powder suction  
flow path, and

5        said gas feed flow path is formed in one gas feed  
pipe constituted by a first portion extending across  
said powder suction flow path and a second portion  
integrally formed with said first portion and  
extending in said powder suction flow path coaxially  
10       to said discharge flow path, said second portion of  
said gas feed pipe having a distal end formed with a  
convergent nozzle separated from said diffuser at a  
predetermined space.

15       33. An apparatus according to claim 32, wherein  
said powder suction flow path is substantially  
straight and cylindrical.

20       34. An apparatus according to one of claims 32  
and 33, wherein a minimum size of a portion around  
said second portion of said gas feed flow path and a  
minimum inner diameter of said discharge flow path are  
set to not less than 10 times a powder particle  
diameter of the powder.

25       35. An apparatus according to any one of claims  
32 to 34, wherein said first portion of said gas feed  
pipe is inclined toward said discharge flow path so  
that an angle thereof with respect to a plane  
perpendicular to the central axis of said powder  
suction flow path is not less than an angle of repose  
of the powder.

36. An apparatus according to any one of claims 32 to 35, wherein said first portion of said gas feed pipe has a circular cross-section.

5 37. An apparatus according to any one of claims 32 to 35, wherein said first portion of said gas feed pipe has a triangular cross-section, and said gas feed pipe is arranged such that a vertex of the triangular cross-section is directed toward an inlet of said powder suction flow path.

10 38. An apparatus according to any one of claims 32 to 37, comprising gas injecting means for injecting a powder removing gas to remove the powder deposited on said gas feed pipe.

15 39. An apparatus according to any one of claims 32 to 38, wherein an inlet portion of said discharge flow path has a conical recessed surface, and an angle of a generating line of said recessed surface with respect to a plane perpendicular to a central axis of said discharge flow path is not less than an angle of  
20 repose of the powder.

40. An apparatus according to any one of claims 32 to 38, wherein an inlet portion of said discharge flow path has a conical recessed surface, and an angle of a generating line of said recessed surface with  
25 respect to a plane perpendicular to a central axis of said discharge flow path is not less than 60°.

41. A powder supply apparatus comprising a first and second inlet means for receiving a powder material and a gaseous material respectively, a discharge means

for discharging said powder material together with  
said gaseous material and a means for generating a  
suction pressure at said first inlet means in response  
to flow of said gaseous material between said second  
5 inlet means and said discharge means whereby the  
powder material is drawn into said flow of gaseous  
material and flow of the gaseous material in the  
direction of said first inlet means is inhibited.

42. A method of supplying a powder material and  
10 a gaseous material to a receiving zone, the method  
comprising the steps of forming a flow of said gaseous  
material between a gas supply means and said reaction  
zone, accelerating said flow at a region intermediate  
said supply means and said receiving zone whereby a  
15 suction pressure is generated, exposing said powder  
material to said suction pressure whereby the powder  
material is drawn into said flow of gaseous material  
and entrained therewith.

43. Any of the powder supply apparatus  
20 substantially as hereinbefore described with reference  
to the accompanying drawings.

44. Any of the methods of supplying powder  
substantially as hereinbefore described with reference  
to the accompanying drawings.



**Patent Act 1977**  
**Examiner's report to the Comptroller under Section 1739 -**  
**(The Search report)**

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**Relevant Technical Fields**

- (i) UK Cl (Ed.M) C3P - PKE, PKJ  
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**Databases (see below)**

(i) UK Patent Office collections of GB, EP, WO and US patent specifications.

Documents considered relevant following a search in respect of Claims :-  
1-44

(ii) ONLINE DATABASES: WPI

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Y: Document indicating lack of inventive step if combined with one or more other documents of the same category. E: Patent document published on or after, but with priority date earlier than, the filing date of the present application.  
A: Document indicating technological background and/or state of the art. &: Member of the same patent family; corresponding document.

Category	Identity of document and relevant passages	Relevant to claim(s)
X Y	US 4993495 (CHEMONICS) see eg Figure 2	1, 41, 42
X Y	US 4915300 (RYAN) see eg the figures	1, 41, 42
Y	US 4687381 (BP) see eg the figures	1

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